AD-A270 232

TATION PAGE

Form Approved
OMB No. 0704-0188

2. REPORT DATE	3. REPORT TYPE AND DATES COVERED
31 August 1993	Final: 1 July 90 to 30 June 93

4. TITLE AND SUBTITLE
Final Technical Report on Grant AFOSR-90-0248
"Tunneling Microscopy of Semiconductors"

5. FUNDING NUMBERS
G-AFOSR-90-0248
Project/Task 2305/A9

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8. PERFORMING ORGANIZATION REPORT NUMBER
AFOSR-TR- 93 0 1 3

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
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Bolling AFB, DC 20332-0001

Program Manager:
Major Billy R. Smith, Jr.
AFOSR/NE

10. SPONSORING/MONITORING AGENCY REPORT NUMBER
2305/A9

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release.
Distribution unlimited.

12b. DISTRIBUTION CODE

13. ABSTRACT *(Maximum 200 words)*

This is the final report of research activities on "Tunneling Microscopy of Semiconductor Surfaces," accomplished during a 3-year JSEP Graduate Fellowship awarded to Silva K. (Leonard) Theiss for work in the Harvard JSEP Laboratory under the guidance of Prof. J. Golovchenko. The work includes: construction of a two-tip tunneling microscope, study of low coverage phases of Pb on Si(111), study of surface diffusion of Pb on Ce(111)-c(2x8), and study of Ge growth on Si(111).

93-23154

14. SUBJECT TERMS	15. NUMBER OF PAGES
Tunneling microscopy, Semiconductor Surfaces, Surface diffusion	3
	16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
Unclassified	Unclassified	Unclassified	UL

NSN 7540-01-280-5500

Standard Form 298 (Rev 2-89)
Prescribed by ANSI Std. 239-18

93 10 1 277

Final Technical Report
JSEP Graduate Fellowship Program
Department of the Air Force
AFOSR - 90 - 0248
Period: 7/1/90 - 6/30/93

Tunneling Microscopy of Semiconductor Surfaces

Silva K. Theiss

Two-tip tunneling microscope.

We constructed a tunneling microscope with two independently-controlled tips in close proximity to each other. We were able make atomic scale images with both tips simultaneously, and to inject a pulse of electrons with one which could be detected with the other. However, because the injected current was small and tended to diffuse in to the bulk sample, our signal-to-noise ratio was not acceptable. Furthermore, we were never able to bring the two tips sufficiently close to each other to do the local conductivity measurements we originally proposed.

Low coverage phases of Pb on Si(111).

I took part in the LEED and tunneling microscopy section of this study, which characterized the phases of Pb on Si(111) from 0 to about 3 monolayers Pb coverage using tunneling microscopy, thermal desorption, Rutherford backscattering, and LEED. One of the main goals was to determine the effect of annealing history on the surface structure, since it had been found that the Schottky barrier height of Pb/Si diodes varied from 0.70 to 0.93 eV on room-temperature-deposited and annealed samples respectively (Heslinga et al., Phys. Rev. Lett. **64**, 1589 (1990).) For room temperature deposition of Pb on Si(111)-7x7 the following phases are observed: 0-0.06ML, Pb adatoms sit above rest atoms with a preference for the faulted half of the unit cell; 0.06-0.12ML, Pb adatoms fill in the remaining rest atom sites; 0.12-0.60ML, Pb and Si adatoms mix to form a 1x1 structure within the boundaries of the 7x7 unit cell; 0.6-1.0ML, Pb atoms fill in the regions above the dimer rows. On annealed samples at low coverage the Pb atoms occupy substitutional sites in the 7x7 reconstruction, at 1/6th ML there is a $\sqrt{3}$ x$\sqrt{3}$ phase which is a 50-50 mix of Pb and Si, at 1/3 ML there is a $\sqrt{3}$ x$\sqrt{3}$ phase which is purely Pb, between 1/3 and 1 ML a 1x1 phase occurs, and above 1 ML there is a rotated incommensurate phase. This work has been published in Surface Science. ("Growth and morphology of Pb on Si(111)," E. Ganz, I.-S. Hwang, F. Xiong, S.K. Theiss, and J.A. Golovchenko, Surf. Sci. **257** 259 (1991).)

Study of surface diffusion of Pb in Ge(111)-c(2x8).

We studied the diffusion of Pb adatoms in substitutional positions in the Ge(111)-c(2x8) reconstruction using the tunneling microscope over a range of temperatures from room temperature to 80°C. This was done by taking a series of images in one area on the

surface, and comparing the positions of Pb atoms in subsequent images. We counted the fraction of atoms which had moved in each image at various temperatures. We found an Arrhenius relation between the movement rate and the inverse temperature. On our initial sample, on which approximately 6% of the adatoms were Pb, we found the activation energy for diffusion is 0.54 ± .03 eV and the effective attempt frequency is about 10^6 s^{-1}. In addition, we found a large fraction of "long jumps" (movements of more than one adatom spacing) and found that the single interchanges were highly anisotropic, with motion along the "2x" axis of the c(2x8) reconstruction strongly favored. This work has been published in Physical Review Letters. ("Direct measurement of diffusion by hot tunneling microscopy: activation energy, anisotropy, and long jumps," E. Ganz, S.K. Theiss, I.-S. Hwang, and J.A. Golovchenko, Phys. Rev. Lett. **68** 1567 (1992).) Subsequently, we measured diffusion on the same surface with a lower coverage of Pb adatoms. In this way we hoped to learn more about the "long jumps" we had observed. Interestingly, we found both a higher activation energy for diffusion and a higher attempt frequency at lower coverage. For the lower coverage sample, on which approximately 4% of the adatoms were Pb, we found $E_D = 0.73 \pm 0.03$ eV and $\nu \sim 10^8$ s^{-1}. Since Pb seems to enhance the mobility of all adatoms on the surface, it is not surprising that there is a lower activation energy for diffusion in the higher coverage case. The reason for the change in the prefactor is not clear. Still, in both cases the measured attempt frequency is orders of magnitude smaller than that usually assumed in a single atom process, which is the Debye frequency, ~ 10^{13} s^{-1}. These results were presented by Professor Golovchenko at the Fall Meeting of the Materials Research Society and are included in the published proceedings. ("Metastable structural surface excitations and concerted adatom motions: A STM study of atom motions within a semiconductor surface." Jene A. Golovchenko, Ing-Shouh Hwang, Eric Ganz, and Silva K. Theiss. MRS Proc. Vol. **295**, 41 (1993).) We also found that for the "long jumps", as with the single interchanges, motion along the "2x" axis of the c(2x8) reconstruction was strongly favored. These data will be included in a paper summarizing our studies of dynamics on the Pb/Ge(111) surface.

Study of Ge growth on Si(111).

1. Construction and calibration of new STM. I modified the second STM in our laboratory from a "louse" design to an "inchworm" design, which is more rigid and thus less susceptible to vibration. The load lock, sample manipulator, electronics, and control software were modified to accommodate the new STM. A Ge effusion cell was added. The instrument was tested in vacuum, and the response of the piezoelectric tube scanner was calibrated using clean Si. Vertical calibration was done using a single bilayer-high step, and horizontal calibration was done using the spacing of the corner holes in the 7x7 reconstruction.

2. Anomalous growth of Ge at low coverages. The new tunneling microscope was used to study the initial stages of Ge growth on Si(111). At 450°C, growth takes place out from the bottom sides of step edges and in small, one- to two- monolayer high islands on the terraces. Enhanced nucleation of islands occurs along the domain boundaries of the 7x7 reconstruction and, surprisingly, along the top sides of step edges. I presented

these results at the Spring APS meeting. I subsequently confirmed that the anomalous step-top islands are not caused by pinning due to impurities at the initial Si step, because the islands are not left behind at the initial step during additional Ge deposition. However, they may be caused by enhanced nucleation at imperfections in the newly grown layer, because we observe a similar result for silicon homoepitaxy under the same temperature and deposition rate conditions, although at somewhat higher coverage. It is not clear why the pattern of enhanced nucleation on the top sides of step edges continues at coverages of several monolayers.

3. Sample preparation and preliminary relaxation study. A procedure for reliably producing clean Si(111)-7x7 surfaces was developed. Ge was deposited on this surface from an effusion cell, and deposition parameters which yielded Ge islands around 50-200Å high were found. These islands had triangular tops with c(2x8) and associated reconstructions (i.e., 2x2 and c(4x2).) If the sample temperature during deposition was too high, the island tops had a 7x7 reconstruction. The lattice parameter on the tops of these islands was measured perpendicular to the "2x" row direction and compared to the 7x7 corner hole spacing measured on clean Si on the other side of the sample. Preliminary results suggest that substantial relaxation of the islands to the Ge lattice constant has already taken place before the islands have large enough tops to yield reliable measurements, but that some compression persists in relatively tall islands. These results need to be confirmed and expanded, and should be ready to be submitted for publication soon.